AMENDMENTS TO THE CLAIMS

Please cancel claims 3, 46 and 49, add claims 99 and 100, and amend claims 1, 37, 41 and 64 without prejudice. The following listing of claims will replace all previous claims and listings in the application.

1. (Currently Amended) A method of preparing a sample for mass spectrometry analysis, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group; and

b) reacting said analyte with a triarylphosphonium labeling reagent having a reactive group capable of reacting with said exposed group to thereby form a triarylphosphonium-linked analyte; wherein said labeling reagent has a structure according to the formula

$[Ar_3P^+R]X^-$

wherein

each Ar is an unsubstituted aryl group, all of which may the same or different;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed group to form a covalent bond thereby forming triarylphosphonium-linked analytes; and

X^- is a negatively-charged counter ion.

2. (Previously Presented) The method of claim 1, wherein the method comprises the further step of obtaining the triarylphosphonium labeling reagent having a reactive group;

3. (Cancelled)

4. (Withdrawn) A method of preparing a sample for mass spectrometry analysis, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group; and

b) reacting said analyte with at least two triarylphosphonium labeling reagents according to the formulae

$[Ar_3P^+R]\ X^-$

and

$[Ar^*_3P^+R]X^-$

wherein

Ar and Ar^* are aryl groups, all of which may the same or different, such that the molecular weight of Ar_3P is different from the molecular weight of Ar^*_3P;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming a triarylphosphonium-linked analyte; and

X^- is a negatively-charged counter ion;

such that at least two triarylphosphonium-linked analytes are formed.

5. (Withdrawn) The method of claim 4, wherein the method comprises the further step of obtaining the at least two triarylphosphonium labeling reagents each having a reactive group, wherein the reactive groups of the labeling reagents are all the same.

6. (Withdrawn) The method of claim 4, wherein the difference in the molecular weights of the triarylphosphonium groups is discernable by mass spectrometry.

7. (Withdrawn) The method of claim 4, wherein the difference in the molecular weights of the triarylphosphonium-linked analytes is discernable by mass spectrometry.

8 (Withdrawn) A method of preparing a sample for mass spectrometry analysis, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group; and

b) reacting said analyte with at least two labeling reagents according to the formulae $[Ar_3P^+R]X^-$

$[Ar^{*}_{3}P^{+}R]X^{-}$

[Ar**3P+R] X^{-}

wherein

the Ar groups (*i.e.*, Ar, Ar^{*}, and Ar^{**}, etc.) are aryl groups, all of which may the same or different, such that the molecular weights of the triarylphosphonium groups of each labeling reagent are unique;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes; and

X^{-} is a negatively-charged counter ion.

9. (Withdrawn) A method of analyzing a sample, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group;

forming a triarylphosphonium-linked analyte by reacting said analyte with a triarylphosphonium labeling reagent having a reactive group that reacts with said exposed group to form a covalent bond thereby forming a triarylphosphonium-linked analyte

such that said triarylphosphonium-linked analyte is formed; and

b) analyzing said triarylphosphonium-linked analyte by mass spectrometry.

10. (Withdrawn) The method of claim 9, wherein the method comprises the further step of obtaining a the triarylphosphonium labeling reagent having a reactive group.

11. (Withdrawn) The method according to claim 10, wherein said labeling reagent has a structure according to the formula

$[Ar_{3}P^{+}R]\ X^{-}$

wherein

each Ar is an aryl group, all of which may the same or different;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed group to form a covalent bond thereby forming triarylphosphonium-linked analytes; and

X^- is a negatively-charged counter ion.

12. (Withdrawn) A method of analyzing a sample, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group; and

b) reacting said analyte with at least two triarylphosphonium labeling reagents according to the formulae $[Ar_3P^+R]X^-$

and

$[Ar^*_3P^+R]X^-$

wherein

Ar and Ar^* are aryl groups, all of which may the same or different, such that the molecular weight of Ar_3P is different from the molecular weight of Ar^*_3P;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming a triarylphosphonium-linked analyte; and

X^- is a negatively-charged counter ion;

such that at least two triarylphosphonium-linked analytes are formed; and

c) analyzing said at least two triarylphosphonium-linked analytes by a mass spectrometry technique.

13. (Withdrawn) The method of claim 12, wherein the method further comprises the step of obtaining the at least two triarylphosphonium labeling reagents each having a reactive group, wherein the reactive groups of the labeling reagents are all the same.

14. (Cancelled)

15. (Withdrawn) The method according to claim 9, wherein said mass spectrometry technique is matrix-assisted laser desorption/ionization mass spectrometry or electrospray mass spectrometry.

16. (Withdrawn) The method according to claim 15, wherein said technique is quantitative.

17. (Withdrawn) The method of claim 13, wherein the step of reacting said analyte with at least two triarylphosphonium labeling reagents

1) reacting, in a first vessel, the first labeling reagent with a first portion of said sample such that triarylphosphonium-linked analytes thereof are formed;

2) reacting, in a second vessel, the second labeling reagent with a second portion of said sample such that triarylphosphonium-linked analytes thereof are formed; and

3) combining triarylphosphonium-linked analytes from said first vessel with triarylphosphonium-linked analytes from said second vessel to form a mixture; and

wherein the step of analyzing comprises analyzing said mixture of triarylphosphonium-linked analytes by a mass spectrometry technique.

18. (Withdrawn) The method of claim 17, further comprising quantitatively comparing the relative signals of the triarylphosphonium-linked analytes from said first vessel to the triarylphosphonium-linked analytes of said second vessel.

19. – 22. (Cancelled)

23. (Original) The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted aryl groups.

24. (Original) The method according to claim 1, wherein each Ar group is selected from the group consisting of substituted or unsubstituted heteroaryl groups.

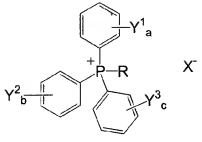
25 – 36. (Cancelled).

37. (Currently Amended) The method according to claim 1, wherein said Ar_3P group is selected from the group consisting of ~~substituted or~~ unsubstituted triphenylphosphine, naphthyldiphenylphosphine, dinaphthylphenylphosphine, trinaphthylphosphine, 9-

anthryldiphenylphosphine, 9-anthryldinaphthylphosphine, diphenylpyrenylphosphine, dinaphthylpyrenylphosphine.

38. – 40. (Cancelled)

41. (Currently Amended) The method according to claim 1, wherein said labeling reagent has a structure according to the formula



wherein

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes;

a, b, and c are ~~independently integers from~~ each 0 ~~to 5~~;

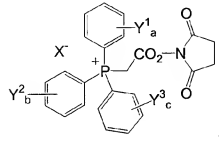
Y^1, Y^2, and Y^3, which may be the same or different, are independently selected from the group consisting of halogen, hydroxyl, alkylcarbonyloxy, arylcarbonyloxy, alkoxycarbonyloxy, aryloxycarbonyloxy, carboxylate, alkylcarbonyl, alkoxycarbonyl, aminocarbonyl, alkylthiocarbonyl, alkoxyl, phosphate, phosphonato, phosphinato, cyano, amino (including alkyl amino, dialkylamino, arylamino, diarylamino, and alkylarylamino), acylamino (including alkylcarbonylamino, arylcarbonylamino, carbamoyl and ureido), amidino, imino, sulfhydryl, alkylthio, arylthio, thiocarboxylate, sulfate, sulfonato, sulfamoyl, sulfonamido, nitro, trifluoromethyl, cyano, azido, heterocyclyl, aralkyl, aryl, and heteroyl groups, provided that none of said Y groups reacts with said R group; and

X^- is a negatively-charged counter ion.

42. (Cancelled)

43. (Cancelled)

44. (Previously Presented) The method according to claim 41, wherein said labeling reagent has a structure according to the formula



45. (Cancelled)

46. (Cancelled)

47. (Cancelled)

48. (Withdrawn) The method according to claim 4, wherein each of said triarylphosphonium labeling reagents has the same chemical structure, and wherein each triarylphosphonium labeling reagent is isotopically enriched with respect to the other triarylphosphonium labeling reagent.

49. (Withdrawn) The method according to claim 48, wherein a triarylphosphonium labeling reagent is isotopically enriched with ^{12}C, ^{13}C, ^{1}H or ^{2}H.

50. (Original) The method according to claim 41, wherein Y^1, Y^2, and Y^3 are selected from the group consisting of $O^{12}C^1H_3$, $O^{12}C^2H_3$, $O^{13}C^1H_3$, and $O^{13}C^2H_3$.

51. (Original) The method according to claim 1, wherein said exposed group of said analyte is electrophilic and said reactive functional group is nucleophilic.

52. (Withdrawn) The method according to claim 1, wherein said exposed group of said analyte is nucleophilic and said reactive functional group is electrophilic.

53. – 55. (Cancelled)

56. (Previously Presented) The method according to claim 3, wherein X^- is a halide, triflate, sulfate, nitrate, hydroxide, carbonate, bicarbonate, acetate, phosphate, oxalate, cyanide, aklylcarboxylate, *N*-hydroxysuccinimide , *N*-hydroxybenzotriazole, alkoxide, thioalkoxide, alkane sulfonyloxy, halogenated alkane sulfonyloxy, arylsulfonyloxy, bisulfate, oxalate,

valerate, oleate, palmitate, stearate, laurate, borate, benzoate, lactate, citrate, maleate, fumarate, succinate, tartrate, naphthylate mesylate, glucoheptonate, or lactobionate.

57. (Previously Presented) The method according to claim 3, wherein X^- is an anionic Y group such that the labeling reagent is zwitterionic.

58. (Withdrawn) A composition comprising at least two different labeling reagents each having a different molecular weight according to the formula

$$[Ar_3P^+R]\ X^-$$

wherein

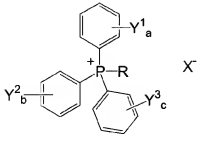
each Ar is aryl group, all of which may the same or different;

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming a triarylphosphonium-linked analyte; and

X^- is a negatively-charged counter ion.

59. (Withdrawn) A composition according to claim 58 comprising at least two different labeling reagents each having a different molecular weight according to the formula



wherein

P is a phosphorous atom;

R is a reactive group comprising a functional group that reacts with said exposed functional group to form a covalent bond thereby forming triarylphosphonium-linked analytes; a, b, and c are independently integers from 0 to 5;

Y^1, Y^2, and Y^3, which may be the same or different, are independently selected from the group consisting of halogen, hydroxyl, alkylcarbonyloxy, arylcarbonyloxy, alkoxycarbonyloxy, aryloxycarbonyloxy, carboxylate, alkylcarbonyl, alkoxycarbonyl, aminocarbonyl, alkylthiocarbonyl, alkoxyl, phosphate, phosphonato, phosphinato, cyano, amino (including alkyl amino, dialkylamino, arylamino, diarylamino, and alkylarylamino), acylamino (including alkylcarbonylamino, arylcarbonylamino, carbamoyl and ureido), amidino, imino, sulfhydryl, alkylthio, arylthio, thiocarboxylate, sulfate, sulfonato, sulfamoyl, sulfonamido, nitro, trifluoromethyl, cyano, azido, heterocyclyl, aralkyl, aryl, and heteroyl groups, provided that none of said Y groups reacts with said R group; and

X^- is a negatively-charged counter ion.

60. (Withdrawn) The composition according to claim 59, wherein each labeling reagent has the same chemical structure, and wherein each labeling reagent is isotopically enriched with respect to the other labeling reagents.

61. – 64. (Cancelled)

65. (Currently Amended) The method according to claim 1, wherein the labeling reagent has the following structure:

wherein

each Ar is unsubstituted aryl group, all of which may be the same or different;

P is a phosphorous atom;

Z is a linking group; and

Ψ is a reactive functional group.

66. (Withdrawn) The method according to claim 65, wherein said reactive functional group is an activated ester of the formula --COL, where L is a leaving group.

67. – 68. (Cancelled)

69. (Cancelled)

70. (Withdrawn) The method according to claim 65, wherein said Ψ group is a carboxylic acid, a derivative of a carboxylic acid, or an activated ester of a carboxylic acid.

71. (Withdrawn) The method according to claim 65, wherein said Ψ group is a haloalkyl, haloacetamide, halomethylbenzamide, a maleimido group, or a sulfonate ester, wherein the sulfonic acid is an alkylsulfonic acid, perfluoroalkylsulfonic acid, or an arylsulfonic acid.

72. (Withdrawn) The method according to claim 65, wherein said Ψ group is an iodoacetamide, maleimide, or a halomethylbenzamide.

73. (Original) The method according to claim 65, wherein said Ψ group is an isocyanate or an acyl nitrile.

74. – 76. (Cancelled)

77. (Original) The method according to claim 65, wherein said Ψ group is an acyl azide, an acyl nitrile, an aldehyde, an alkyl halide, an amine, an anhydride, an aniline, an aryl halide, an azide, an aziridine, a boronate, a carboxylic acid, a diazoalkane, a haloacetamide, a hydrazine, an imido ester, an isocyanate, an isothiocyanate, a maleimide, a sulfonyl halide, or a thiol group.

78. (Cancelled)

79. (Previously Presented) The method according to claim 65, wherein Z has 1-20 nonhydrogen atoms selected from the group consisting of C, N, O and S, and the longest linear segment contains 1-6 nonhydrogen atoms.

80. – 82. (Cancelled)

83. (Original) The method of claim 1, wherein said analyte is a protein, peptide, enzyme, immunoglobulin, hapten, antigen, amino acid, hormone, receptor, nucleic acid, hormone, chemical, polymer, pathogen, toxin, saccharide or polysaccharide, steroid, vitamin, therapeutic drug, drug of abuse, bacterium or virus, or a combination or fragment of any of the foregoing, or a metabolite thereof, or an antibody thereto.

84. (Withdrawn) The method of claim 1, wherein said analyte is a food additive, agrichemical, surfactants, adhesives, resin, organic pollutant, or process chemical.

85. (Withdrawn) The method of claim 1, wherein said analyte is a therapeutic drug or a metabolite thereof.

86. (Withdrawn) The method of claim 1, wherein said analyte is a drug of abuse or a metabolite thereof.

87. (Withdrawn) The method of claim 1, wherein said sample is rainwater, or water from an ocean, river, lake, pond, or stream.

88. (Original) The method of claim 1, wherein said sample is a biological tissue.

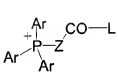
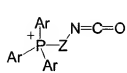
89. (Cancelled)

90. (Withdrawn) A kit for use in preparing a sample for mass spectrometry analysis comprising a triarylphosphonium labeling reagent having a reactive group, and instructions for use in the method of the instant invention.

91. (Withdrawn) The kit according to claim 90 further comprising buffer chemicals.

92. (Cancelled)

93. (Withdrawn) A labeling reagent having a structure selected from the group consisting of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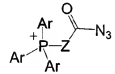
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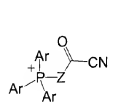
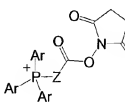
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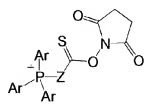
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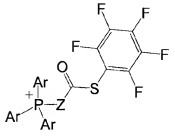
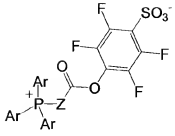
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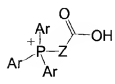
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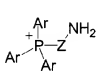
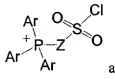
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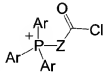
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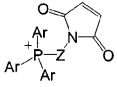
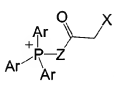
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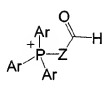
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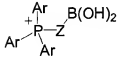
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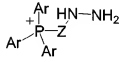
;

wherein

each Ar is aryl group, all of which may the same or different;

P is a phosphorous atom;

Z is a linking group; and

L and X are, independently, leaving groups.

94. – 98. (Cancelled)

99. (New) A method of preparing a sample for mass spectrometry analysis, comprising

a) obtaining a sample comprising an analyte, wherein said analyte comprises an exposed group; and

b) reacting said analyte with a triarylphosphonium labeling reagent having a reactive group capable of reacting with said exposed group to thereby form a triarylphosphonium-linked analyte;

wherein said sample is biological tissue.

100. (New) The method of preparing a sample for mass spectrometry analysis of claim 99 wherein said analyte is a small molecule.